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AMERICAN OPTICAL COMPANY

Southbridge, Massachusetts

NEODYMIUM LASER GLASS
IMPROVEMENT PROGRAM

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1. INTRODUCTION

This report is a technical summary covering work performed in the period between 30 June 1964 and 1 January 1965 on Contract Nonr 3835(00) entitled Laser Materials Research and Development. During this period the main area of investigation has been concerned with athermalizing laser glass. The effects of composition on the expansion coefficient, the thermal coefficient of the index of refraction, the birefringence and the stress-optical coefficients are being studied for this purpose.

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2. TECHNICAL DISCUSSION

2.1 ATHERMALIZATION

In order to obtain diffraction limited beams from laser devices it is not only necessary to have good optical quality glass but also to have no distortion generated in the laser cavity during the pumping process. A distortion does usually occur, however, due to a pump induced non-uniform temperature distribution. With a proper resonant cavity design and a properly chosen glass composition athermalized laser systems, i.e. those which are free of thermally induced distortion, appear to be feasible.

For optically pumped laser rods, changes in the optical path length of the cavity may arise from three factors. First, a pump induced non-uniform change in temperature leads to an elongation of the rod as governed by the coefficient of thermal expansion, α, of the glass. Second, this non-uniform temperature distribution will also cause a non-uniform change in the index of refraction due to the thermal coefficient of the index of refraction, α_n. Finally, thermal gradients in the glass produce stress which results in index changes and birefringence due to the stress optical coefficient of the glass, B.

Morey[1] points out that the parameter commonly referred to as the stress-optical coefficient is actually the difference, at any given point, between the stress-optical coefficients for light polarized at right angles to, and the light polarized parallel to the force producing the stress as viewed from a mutually perpendicular direction. This parameter more accurately describes the birefringence produced by stress and it will therefore be referred to here as the "stress birefringence" coefficient, ΔB. The term stress-optical coefficient will be reserved for the change in index between the stressed and unstressed states as indicated below,

$$n_z - n = B_{\perp} P_y \quad \text{and} \quad n_y - n = B_{\parallel} P_y,$$

where P_y is a thrust in the y direction $B_{\perp}$ and $B_{\parallel}$ are the stress optical coefficients in directions perpendicular to, and parallel to the direction of thrust respectively.

The general approach to the problem of athermalizing laser glass is to derive an expression for the change in the optical path length for various rays and polarizations in the cavity in terms of temperature dependent parameters. Since the stress-optical coefficient is a function of the plane of polarization of the light, expressions must be derived for the changes in path length in both the radial and tangential planes of polarization. Unlike the coefficients of index and expansion which are functions of temperature at the point under consideration, the stress-optical coefficient is a function of stress which may arise from temperature changes in adjacent areas and therefore the physical shape of the laser rod and its overall temperature distribution must be considered.

In the original analysis of this problem by Snitzer of American Optical Company, the effects of expansion, index changes and stress on the optical path length of two extreme physical configurations were considered. In one case the ratio of length, L, to radius, a, is much greater than unity and the condition of plane stress exists, i.e., the stress exists only in a plane perpendicular to the axis of the rod. In the other case the ratio L/a is much less than unity and the condition of plane strain exists. Assuming a radial temperature distribution and choosing an appropriate value of the stress-optical coefficient from the literature (that is, the stress birefringence as defined above) the range of values for α and α_n, which produce zero path difference between the center and the edge of the rod, were determined for these extreme configurations. All other configurations would fall between these extremes and require intermediate values of these parameters.

Subsequently, a more rigorous analysis was carried out by Quelle[2] of ONR for the case of $L/a > 1$ with a cylindrical temperature distribution. It includes the distinction between stress birefringence, ΔB, and the stress-optical coefficients $B_\perp$ and $B_{\parallel}$. From his analysis one obtains the following equations for the radially polarized and tangential polarized differences in path lengths $\Delta P_r(r)$ and $\Delta P_\theta(r)$ for beams of light a distance r from the axis as compared with the ray through the axis where E is Young's modulus, σ is Poisson's ratio and $T(r)$ is the temperature distribution.

$$\Delta P_r(r) = L \left\{ \left[n\alpha_n + \frac{\alpha E}{1-\sigma} \; (2 \; B_\perp) \right] T(r) \right.$$

$$+ \frac{\alpha E}{1-\sigma} \; \left[B_{||} - B_\perp \right] \frac{1}{r^2} \int_0^r T(r) \; rdr$$

$$\left. + \left[2\alpha(n-1) - \frac{\alpha E}{1-\sigma} \; (3 \; B_\perp + B_{||}) \right] \frac{1}{A^2} \int_0^A T(r) \; rdr \right\} \qquad (1)$$

$$\Delta P_\theta(r) = L \left\{ \left[n\alpha_n + \frac{\alpha E}{1-\sigma} \; (B_{||} + B_\perp) \right] T(r) \right.$$

$$+ \frac{\alpha E}{1-\sigma} \; \left[B_\perp - B_{||} \right] \frac{1}{r^2} \int_0^r T(r) \; rdr$$

$$\left. + \left[2\alpha(n-1) - \frac{\alpha E}{1-\sigma} \; (3 \; B_\perp + B_{||}) \right] \frac{1}{A^2} \int_0^A T(r) \; rdr \right\} \qquad (2)$$

In both Eqs. (1) and (2) only the first two terms are radius dependent. The third term is therefore only capable of introducing changes in path length which will result in a change in the wavelength of the laser radiation and will not distort a plane wave passing through the laser system. The athermalization of the glass is feasible only because, with changes in composition of the glass, the value of the thermal coefficient of the index of refraction at 1.06 μ may be varied from about $-40 \times 10^{-7}/C°$ to about $+40 \times 10^{-7}/°C$.

The conditions necessary for athermalization, i.e., $\Delta P = 0$, are thus determined by the first two terms of Eqs. (1) and (2). In general to make ΔP equal zero the quantities in the square brackets must be made equal to zero. Two general cases exist, the first in which $B_\perp = B_{||}$, that is zero stress-birefringence, the second in which $B_\perp \neq B_{||}$.

<u>CASE I</u>

If $B_\perp = B_{||}$ the second terms in Eqs. (1) and (2) vanish since the quantity within the square bracket, ΔB, equals zero. In addition, the first terms become identical and the change in path length is the same for both directions of polarization as prescribed for the case of zero birefringence. In order to make the ΔP's equal zero the quantity within the square brackets in the first terms must equal zero, i.e.

$$n\alpha_n + \frac{\alpha E}{1-\sigma} \; (2 \; B) = 0. \qquad (3)$$

The value of α_n must be a negative quantity since the other parameters in the equation are positive and σ is less than unity.

At the present time the only glass known to have zero stress-birefringence is one introduced by Pockels[3] containing about 76 wt.% lead oxide. Unfortunately, this glass has a positive value of α_n at room temperature and above and therefore Eq. (3) cannot be satisfied. Lowering the temperature shifts α_n toward more negative values but there is insufficient data at the present time to tell if the shift would be sufficient in this case. One of our present goals is to determine the effect on ΔB of the various oxides used in making glass so that the feasibility of developing other zero stress-birefringence glasses may be determined.

CASE II

When $B_\perp$ and $B_{||}$ are not equal the glass exhibits stress-birefringence and the values of $\Delta P_r(r)$ and $\Delta P_\theta(r)$ are never equal as may be seen from Eqs. (1) and (2). It is no longer possible to choose a set of parameters for Eqs. (1) and (2) such that ΔP_r and ΔP_θ are both zero. Two alternative methods of athermalization are therefore being considered in which the path length for a ray near the edge of the laser is made equal to the path length along the center of the rod.

The first method is to choose the parameters such that the average path length for the two directions of polarization near the edge of the rod is equal to the path length at the center as shown in Eq. (4)

$$\frac{\Delta P_r(r) + \Delta P_\theta(r)}{2} = \frac{L}{2}\left[2n\alpha_n + \frac{\alpha E}{1-\sigma}\left(B_{||} + 3\,B_\perp\right)\right]T(r) = 0 \qquad (4)$$

A cavity design is chosen such that the direction of polarization, for a ray oscillating within the cavity, alternates between radial and tangential for each successive passage through the active laser element. Two such systems are shown in Figure 1 (a and b). In Figure 1a, a 45° Faraday rotator is interposed between the laser rod and the totally reflecting end mirror. Thus a tangentially polarized ray will become a radially polarized ray in the process of being totally reflected at this end of the cavity and vice versa. This results in a path length at the edge of the rod which is the average of the optical path lengths of these two planes of polarization as the light passes back and forth between end reflectors. A similar result can be achieved in a cascaded system as shown in Figure 1b by introducing a 90° Faraday rotator between the laser components.

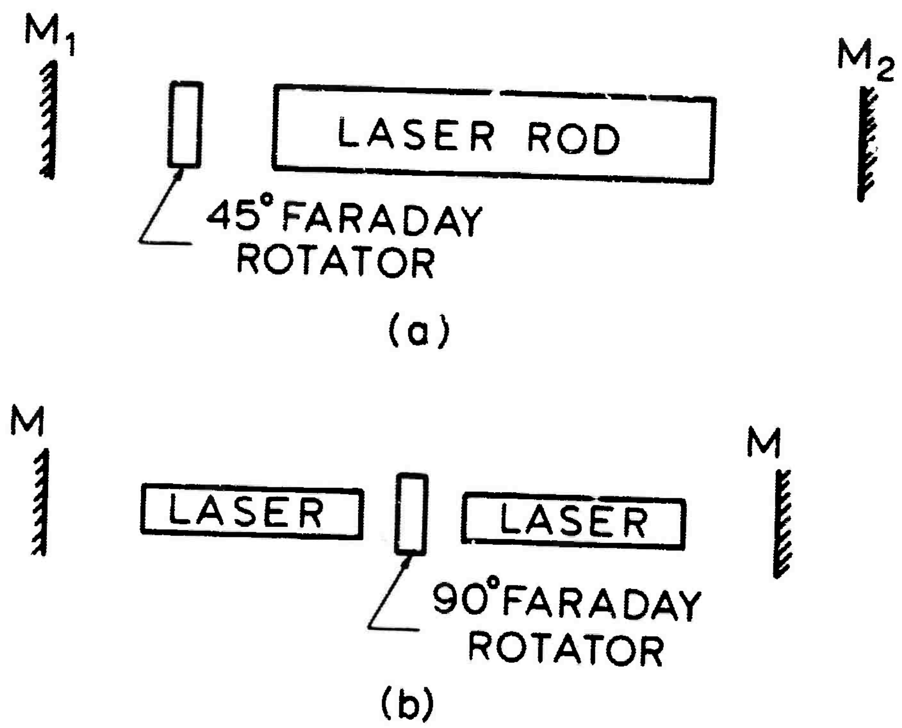

Figure 1. Schematic Diagrams of Lasers with Compensating Faraday Rotators

The second method of making the path length at the edge of the rod equal to that at the center, for the case where $B_\perp$ and $B_{\parallel}$ are not equal, is as follows. The edge and center path lengths are made equal either for rays which are tangentially polarized or for those which are radially polarized, but not both. The cavity is then operated only in that mode for which the path lengths are equal, i.e., if the path lengths are equal for radially polarized light then the radially polarized TM_{01} mode is excited and vice versa.

3. EXPERIMENTAL PROGRAM

In the above schemes for athermalization we state that parameters may be chosen to satisfy a certain set of conditions. The goal of this project is to provide sufficient latitude in the parameters involved to make this possible. It is desirable therefore to know the available range of values of parameters such as n, α, α_n, $B_\perp$, $B_{\parallel}$, and ΔB, and to know how the values within the range vary with composition.

For this purpose a series of glass compositions has been designed which includes the more commonly used glass ingredients. Each oxide was used in at least four different concentrations covering the range exceeding that encountered in normal glass technology. Individual glasses contained from six to nine oxides. Since the resulting data were to be submitted to a regression analysis, care was taken to be sure that no cross-correlation in composition existed between glasses, that is, that the ratio of concentrations of any two ingredients in one glass is not repeated in any other glass. A second series of glasses has been added to broaden our scope, which is similar to the above, but contains less commonly used ingredients.

A knowledge of the effect of these glass ingredients on the above parameters should provide the flexibility required for the athermalization schemes previously suggested. To acquire this information the general procedure is to collect enough data to make the results significant and then subject it to a regression analysis. With the limited number of glasses involved in the present case, this requires that almost all of the data be available before the analysis is made.

It should be borne in mind that some parameters may not have a simple dependence upon composition, i.e. that the regression equation may be complex and thus more difficult to determine. Our experience in the past, however, indicates that this technique is a powerful tool for determining trends which are composition dependent.

3.1 THERMAL COEFFICIENT OF INDEX OF REFRACTION

The method of measuring α_n, used in this work takes advantage of the spectral characteristics of the output of a glass laser operating under special conditions; namely, the laser may be made to emit a spectrum of light which consists of a series of sharp lines equally spaced over a wavelength range limited by the fluorescent linewidth of the active ion. This is done by incorporating a thin plate of transparent material in the resonant cavity as the partially transmitting end reflector. The reflectivity of the plate is increased to somewhat less than 16% for wavelengths at which the optical thickness of the plate, nL, is equal to an odd number of quarter wavelengths, whereas the Fresnel reflection normally encountered at a glass air interface is 4%. The wavelengths λ at which laser action occurs is thus governed by the physical thickness L and the index of refraction n of the glass plate, N being an integer, as shown in Eq. (5).

$$nL = (2N + 1)\ \lambda/4 \tag{5}$$

When the temperature is changed both the physical thickness and the index of refraction of the glass plate will change. Thus a change in the temperature of the glass plate will result in a shift in the wavelengths at which laser action takes place. Equation (5) thus leads to the following expression for the shift in wavelength with respect to temperature.

$$dL/Ldt + dn/ndT = d\lambda/\lambda dT \tag{6}$$

$$\text{or} \qquad \alpha + \alpha_n = \alpha_T \tag{7}$$

The two terms on the left hand side of Eq. (6) are the expansion coefficient, α, and the thermal coefficient of the index of refraction, α_n. The measurement of α_T, the shift in wavelength of the laser emission as a function of temperature, plus an independent measurement of the expansion coefficient of the glass plate used as the partial reflector thus enables one to determine the thermal coefficient of the index of refraction of this plate.

The apparatus for measuring the wavelength shift as a function of temperature is shown schematically in Figure 2. A clad neodymium glass laser 1/4 inch diameter and 18 inches long was used for this work. The cladding has an index of refraction only slightly lower than the core glass which results in a beamspread of about 15°. The high reflecting end of the rod is a 90° roof to provide essentially 100% total internal reflection. The exit end of the rod is just a polished flat surface with a nominal 4% Fresnel reflection.

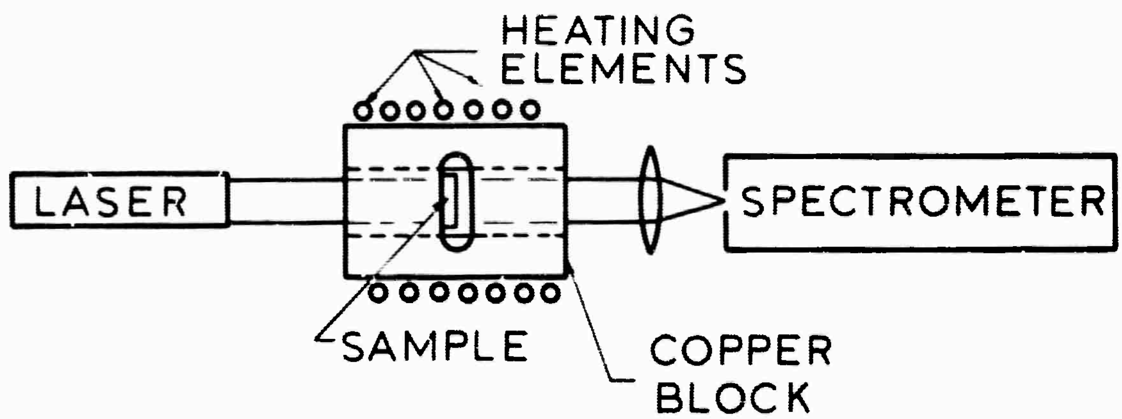

Figure 2. Schematic Diagram of Apparatus for Measuring Thermal Coefficient of the Index of Refraction

Reflection plates about 1/2 millimeter thick were cut from the glasses to be studied. The plates were polished to an optical parallelism of 5 fringes or less per centimeter. They are placed in a block of copper, for thermal stability, which is equipped with a thermocouple and a heating element to provide the required temperature increments. The light from this system is focused upon the slit of a model #70-320 Jarrell-Ash grating spectrometer with a 3.4 meter Ebert mount. The laser is operated near threshold to produce sharp emission lines and to prevent damage to the spectrometer slits. Spectra are recorded on infrared sensitive Kodak type 1-Z spectroscopic plates.

Spectra of a typical glass sample as a function of temperature are shown in Figure 3. This photograph illustrates how all the lines shift in wavelength with temperature. From this data a curve of $\Delta\lambda/\lambda$ vs. T may be plotted, the slope of which is α_T. A gradual increase in α_T is observed which is probably due to changes in α and/or α_n with temperature, but may be due in part to experimental error.

Care must be taken in supporting the glass sample in the copper block to prevent the possibility of introducing error due to stress in the sample produced by the sample support. The distance between the sample and the end of the laser rod is set so that light reflected from the sample is essentially integrated over a larger fraction of the area. This was necessary since the optical thickness of the samples may vary slightly from point to point over the surface due to stria and therefore any slight movement of the sample as the copper block expands would appear as a change in thickness.

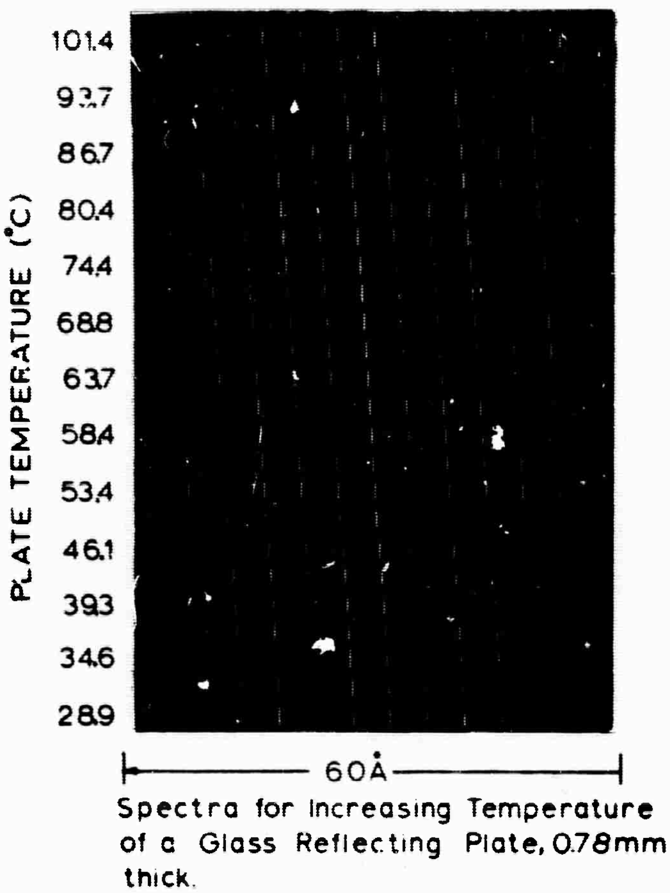

Figure 3. Spectral Output of a Glass Laser with a Thin Plate Acting as a Reflection Filter for Various Temperatures of the Plate

The present system appears to be quite insensitive to motion of the glass sample. The spectra in Figure 4 were made at constant temperature to show the effect of rod to sample distance on the degree of wavelength shift introduced by controlled movement of the sample. These data indicate that this system can provide α_n values with a high degree of reproducibility. The data will be analyzed to determine the effect of the individual glass ingredients on α_n. A direct measurement of α_n from the visible to 1.06 μ is in progress as a check on the accuracy of the above technique.

3.2 COEFFICIENT OF THERMAL EXPANSION

The value of the expansion coefficient used in the calculation of α_n has been determined with a Chevenard type dilatometer. An accurate value of α is desired because the value of α can be several

times larger than α_n. The percentage error in α_n will therefore be correspondingly larger than the error of α due to the above method of calculating α_n.

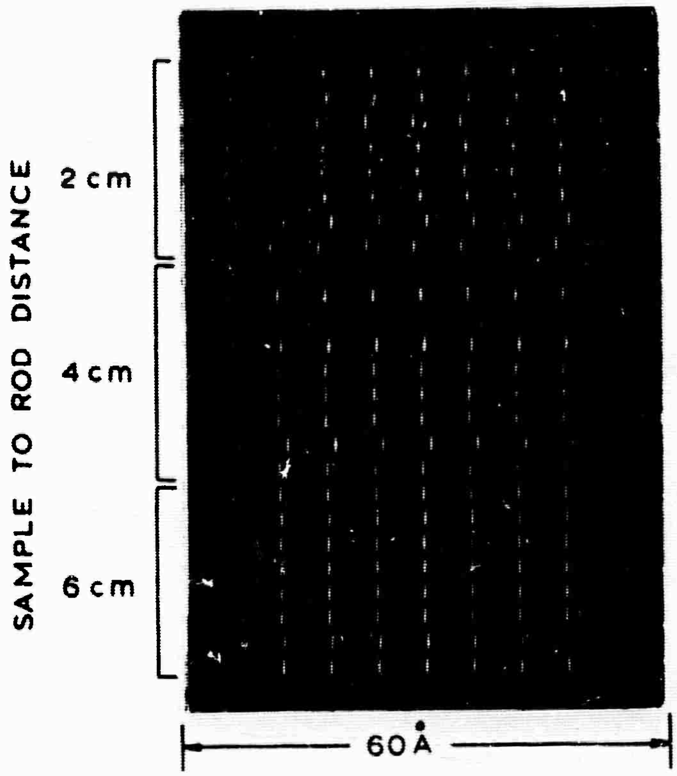

Figure 4. Wavelength Shift Produced by Controlled Movement of the Sample Plate as a Function of Distance between the Plate and the End of the Laser Rod. Movements in each sequence include rotation about a horizontal axis of $+1/5°$ and $-1/5°$, horizontal displacement of +0.1 mm and -0.1 mm, and rotation about a vertical axis of $+1/5°$ and $-1/5°$.

Converting the dilatometer trace into a plot of dL/L vs. T, results in an error in the individual determinations of $\Delta L/L$ of $1\frac{1}{2}$ to 3%. The possibility of measuring α by a more accurate interferometric method is being considered. Careful expansion measurements by Kishii[4] show that the curve over the range 25 to 125°C usually shows a very slight increase in rate of expansion as temperature increases. This provides additional impetus for a more accurate determination of the expansion curve, for a few selected glasses at least. It would appear that the contribution of the temperature dependence of the expansion coefficient to the shape of the α_t curve will probably be small.

A regression analysis program has been set up for an IBM 1620 computer to determine the regression coefficients of the various glass ingredients for the expansion coefficient. The equation used in this analysis is

$$\alpha = b_1 x_1 + b_2 x_2 + \dots . b_n x_n$$

where b_i is the regression coefficient and x_i is the weight percent of the i^{th} oxide in the composition. In Table I are listed some regression coefficients taken from the work of Hall[5] which he arrived at "by inspection and trial" compared with values obtained from some of the same data by our computer analysis. In addition some preliminary values obtained from our own data are given. The agreement between the two methods using the same data is quite good, indicating that the methods themselves are fairly accurate. Analysis of the other oxides in the series will be performed as soon as enough data is available to make the results meaningful.

TABLE I Regression Coefficient

OXIDE	HALL	IBM 1620 OF HALL DATA	IBM 1620 AO DATA
SiC_2	*	.02	.01
Li_2O			.54
Na_2O	.38	.44	.46
K_2O	.30	.34	.42
Rb_2O			.24
CaO	.15	.16	.21
BaO	.12	.13	.12
MgO	.02	.03	
ZnO	.10	.10	
B_2C_3	.02	.03	
Al_2O_3	.05	.08	-.05
PbO	.075	.09	.107

*non-linear

3.3 STRESS-OPTICAL COEFFICIENT

As previously stated, data is being taken for an analysis of ΔB as a function of composition in an attempt to develop glasses, other than the present Pockels glass, which will have zero birefringence.

The stress-birefringence is measured using a system originally described by Cornu[6] in which a rod of glass with rectangular cross section is flexed over a double fulcrum system. The measurement

of retardation due to stress is made between the two fulcrum points using a quartz wedge. This apparatus also may be used to determine Poisson's ratio, σ, and Young's modulus, E.

The system shown in Figure 5 has been set up to measure the stress optical coefficients, $B_{\perp}$ and $B_{\parallel}$. A cubical sample, mounted in series with a Dillon Force gauge between the jaws of an arbor press, is located in one arm of a Michelson interferometer. With the aid of a polarizing element it is possible to measure the actual change in path length, for a given applied pressure, both parallel to, and perpendicular to the direction of thrust. If one face of the sample, which may be rotated, is silvered then this may be used as one mirror of the interferometer and the actual deformation of the sample due to the applied pressure may be observed. This will provide a means of distinguishing how much of the change in optical path length is due to a change in index and how much is due to a change in physical dimensions.

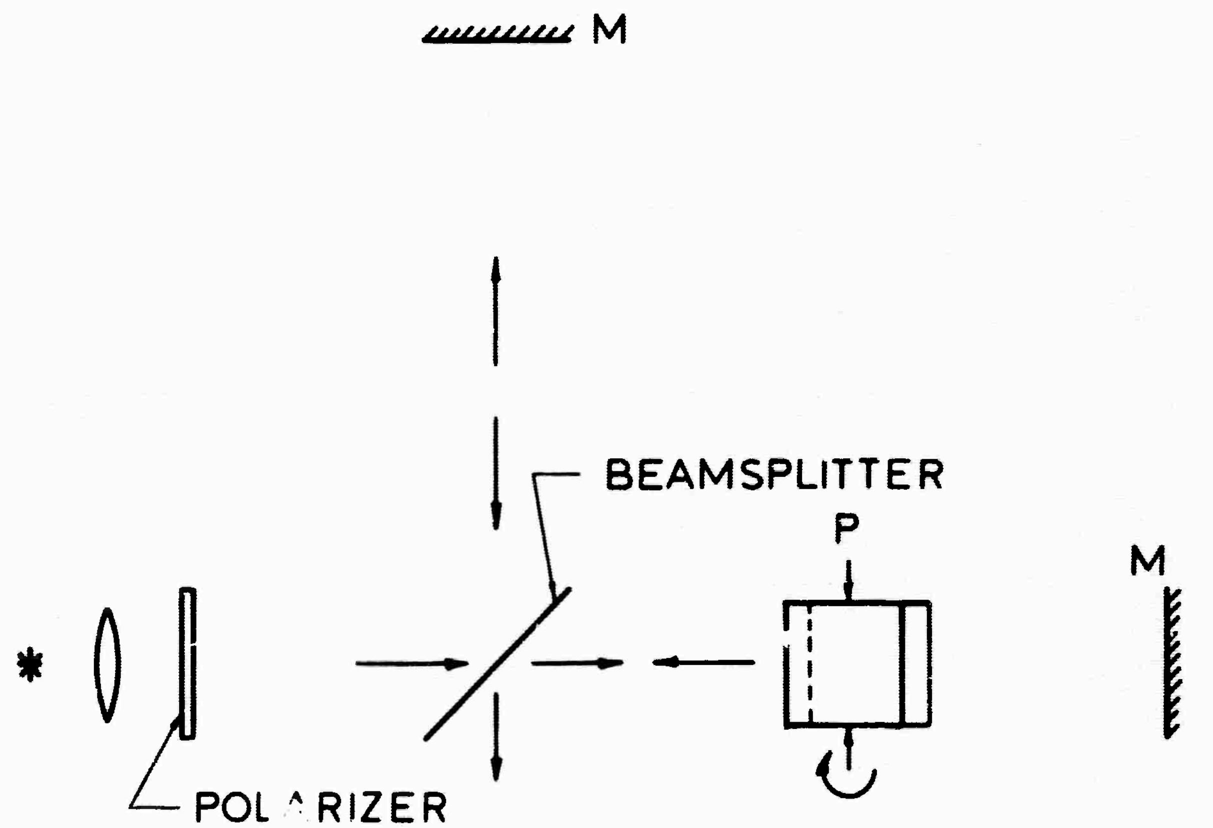

Figure 5. Schematic Diagram of Interferometer System for Measuring $B_{\perp}$ and $B_{\parallel}$

4. SUMMARY

In summary, it is felt that an athermalized laser system can be produced by counterbalancing the index of refraction changes produced by stress with index changes produced by temperature directly. Systems using both Pockels type and non-Pockels type glasses have been considered. The study of parameters such as n, α, α_n, $B_\perp$, $B_\parallel$, and ΔB as a function of composition, which makes athermalization feasible, is now in progress.

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